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Supporting Information

Efficient Removal of Short-Chain and Long-Chain PFAS by Cationic Nanocellulose

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Text S1. Fourier-Transform Infrared Spectroscopy (FTIR)

ALPHA FT-IR Spectrometer (Bruker Optics Inc.) was used to detect the functional groups of wood pulp and QNC samples over the wavelength range of 400-4000 cm^{-1}s. The attenuated total reflectance (ATR) mode was used to study the samples in the solid form.

Text S2. X-Ray Diffraction (XRD)

XRD patterns of varying samples were measured by a Rigaku MiniFlex 600 Benchtop X-ray Diffractometer with diffraction angle from $5° \leq 2\theta \leq 40°$ at a scanned rate of $10°min^{-1}$. The instrument was powered by the Cu Kα radiation (λ = 0.154 nm through a Ni filter) generated at 40 kV and 40 mA. The crystallinity index (CI) was calculated as the ratio between the area of the crystalline peaks (A_{cr})and the total area (A_{total}) under the diffraction profile, as expressed by the following equation:

$$\mathrm{CI} = \frac{A_{cr}}{A_{total}} \times 100\% \quad (1)$$

Individual crystalline peaks were extracted by the peak deconvolution method. The chosen peak fitting program was developed in Python, which utilized Lorentzian functions to fit each crystalline peak. A broad background peak attributed to the amorphous phase centered around 21.5° (1.52 $Å^{-1}$) was also fitted with a Gaussian function.[1] The XRD data indicated the all observed crystal structure was cellulose I.[2]

Text S3. Conductivity Titration

Conductivity titration on the QNC suspension was carried out as follows. About 100 mL of 0.01 M silver nitrate solution was prepared prior the titration. Subsequently, 50 mL of

0.03 wt% QNC suspension was stirred for 10 min and was titrated with 0.5 mL silver nitrate at a 30 s interval. The number of quaternary ammonium groups equal to the chloride ions was determined by the volume of silver nitrate used for each sample.[3]

Text S4. Zeta Potential Measurement

A Zetaprobe Analyzer (Colloid Dynamics, LLC) was used to measure the zeta potential of wood pulp and QNC samples. The probe was first washed and calibrated using the standard zeta probe polar solution (KSiW 0.26 mS/m solution). Then around 280 mL of 0.1 wt% QNC suspension was filled in the sample holder to conduct 10 consecutive measurements under magnetic stirring. The reported zeta potential value was based on the average number of 10 measurements at pH = 6-7. For the measurement under different pH condition, 0.1 mol of HCl and NaOH was used to adjust the pH level from 2-10.

Text S5. Surface Area Measurement

The surface area of QNC samples was determined by the N_2 adsorption isotherm curve using Quantachrome NOVAtouch LX2 analyzer, equipped with degasser and Brunauer–Emmett–Teller (BET) analyzer units. Before the measurement, the sample went through the degassing process at 100 °C (initiated at a heated rate of 10 °C/min) for 12 h under a dry N_2 gas flow. The full BET isotherm measurement (0.05 – 0.99 P/P_o, where P/P_o represents the relative pressure) was performed under helium using the fine powder model, where the specific surface area was analyzed by applying the multi-points BET adsorption−desorption isotherm model in the range of 0.05 – 0.35 P/P_o.

Text S6. Electron Microscopy

To study the morphology and fiber size of the pristine cellulose and QNC, scanning electron microscope (SEM) (Hitachi 4800, Japan) and transmission electron microscope (TEM) (JEOL 1400 LaB6 Soft Bio TEM, Japan) were used, respectively. Air dried pristine cellulose and freeze-dried QNC were selected for SEM analysis. For TEM measurements, samples were prepared by using 0.005% concentration and 10 µL of QNC suspension, casted on copper grids (300 mesh, Ted Pella Inc.). All TEM samples were stained using a 0.2 µL of 2.0 wt% uranyl acetate solution.

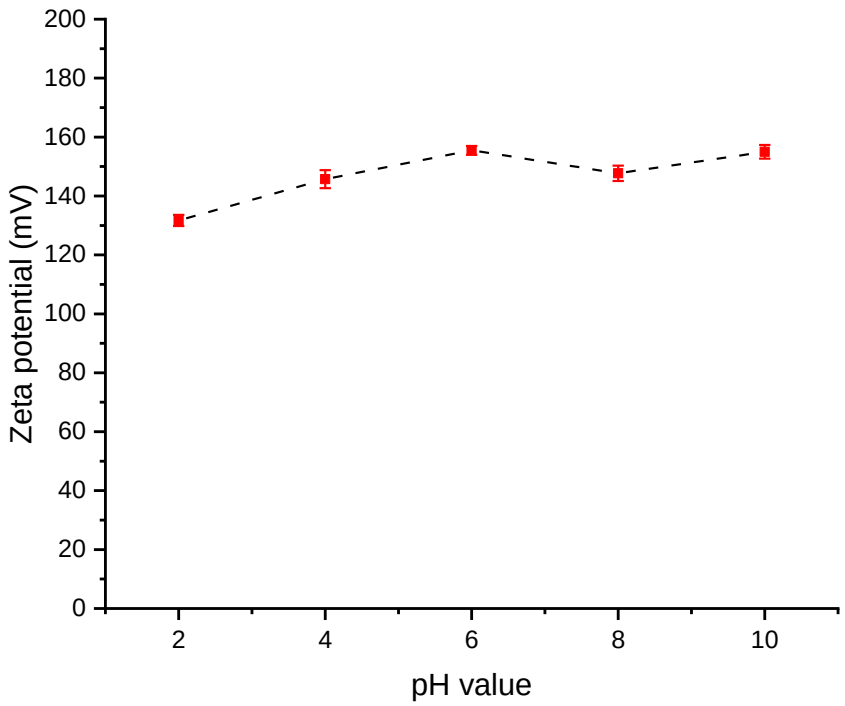

Figure S1. Zeta potential values for QNC 12:1 under different pH values.

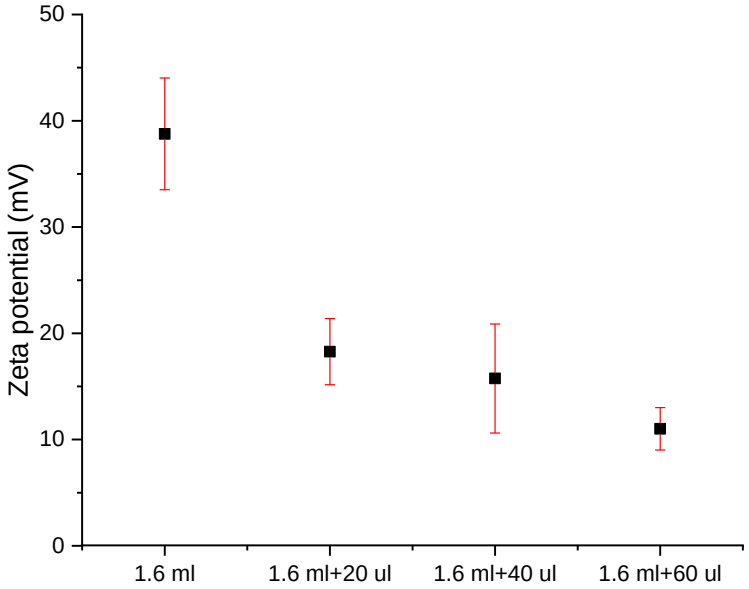

Figure S2. Zeta potential values for QNC 12:1 after PFOA adsorption. (1.6 mL 0.03 wt% of QNC plus 20, 40 and 60 µl of 1g/L PFOA)

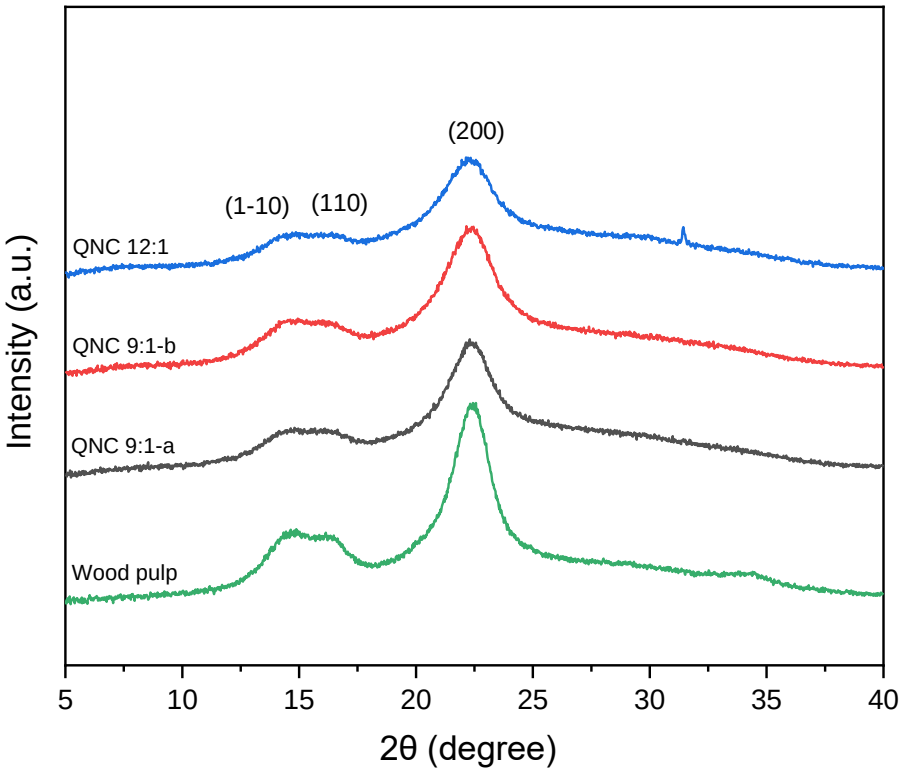

Figure S3. XRD profiles for wood pulp, QNC 9:1-a, QNC 9:1-b and QNC 12:1.

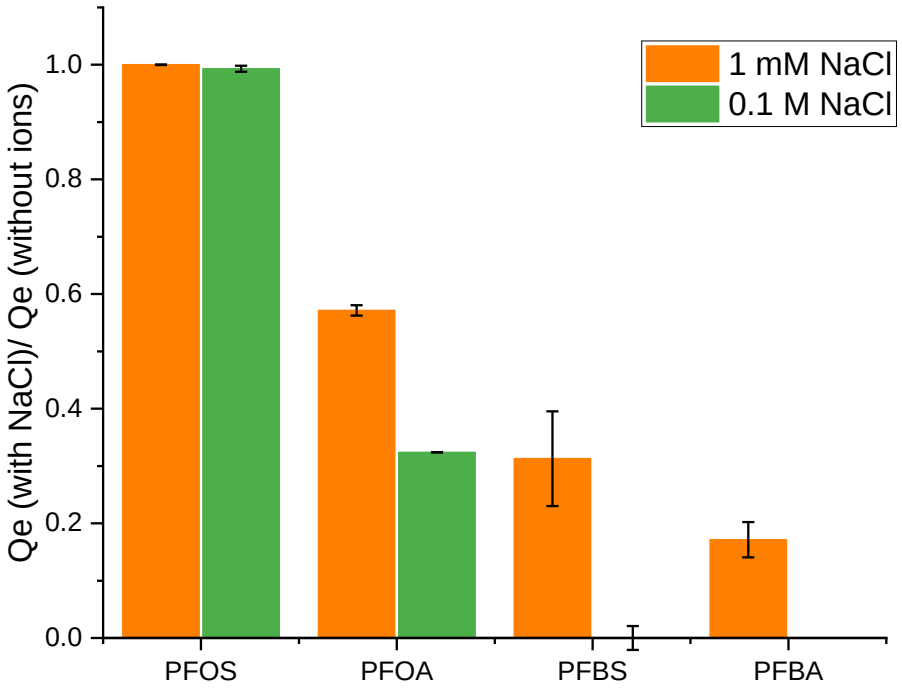

Figure S4. NaCl ion test for four different PFASs on QNC. (For PFOS/PFOA, C_o = 2 ppm, QNC = 32 mg/L; for PFBS/PFBA, C_o = 10 ppm, QNC = 320 mg/L

Table S1. Basic information for the ground water sample in Long Island, NY

Water quality parameter	**Long Island groundwater**
Conductivity (µS/cm)	25.8
ORP (mV)	117
Chloride (mg/L)	7.3
Sulfate (mg/L)	6.09
Total Organic Carbon (mg C/L)	2.59
Ammonia (mg N/L)	0.03
TKN (mg N/L)	BDL
Nitrate + Nitrite (mg N/L)	0.57
Nitrite (mg N/L)	0.01
Orthophosphate (mg P/L)	BDL

Table S2. The PFAS quantitatively analysis results using the ground water (GW) samples.

PFAS Compound	GW Back-ground (μg/L)	GW with 32 mg/L Gel (μg/L)	GW with 80 mg/L Gel (μg/L)	GW with 160 mg/L Gel (μg/L)	GW with 320 mg/L Gel (μg/L)	GW with 640 mg/L Gel (μg/L)
PFBA (C3)	0.153	0.142	0.149	0.153	0.168	0.169
PFPeA (C4)	0.321	0.301	0.300	0.299	0.316	0.298
PFBS (C4)	0.675	0.589	0.588	0.477	0.513	0.396
4-2FTS (C4)	0.037	0.032	0.028	0.026	0.025	0.020
PFHxA (C5)	1.357	1.130	1.087	0.955	0.937	0.704
PFPeS (C5)	0.736	0.360	0.281	0.134	0.061	0.035
PFHpA (C6)	0.184	0.073	0.063	0.031	**<0.010**	**<0.010**
PFHxS-total (C6)	3.449	0.200	0.172	0.059	0.040	**<0.010**
6-2FTS (C6)	0.891	0.036	0.034	0.012	**<0.010**	**<0.010**
PFOA (C7)	0.211	**<0.010**	**<0.010**	**<0.010**	**<0.010**	**<0.010**
PFHpS (C7)	0.094	**<0.010**	**<0.010**	**<0.010**	**<0.010**	**<0.010**
PFOS-total (C8)	3.578	0.032	0.028	0.029	0.030	0.029
8-2FTS (C8)	0.038	**<0.010**	**<0.010**	**<0.010**	**<0.010**	**<0.010**

*The bold face number means the value was below the detection limit of the LC-MS instrument as 0.01 μg/L.

Table S3. LC-MS/MS operating conditions and corresponding parameters.

LC Experimental Conditions	
Parameter	*Value*
LC	Agilent G7120A 1290 Binary Pump Agilent G7116A 1260 Multi-column Thermostat Agilent G7167A 1260 Multi-sampler
Analytical Column	Agilent ZOBRAX Eclipse Plus C18 3.0 x 50 mm, 1.8 μm
Delayed Column	Agilent ZOBRAX Eclipse Plus C18 4.6 x 50 mm, 3.5 μm
Column Temperature	50 °C
Injection Volume	5 μL
Mobile Phase	(A) 5 mM Ammonium Acetate in Water B() 100% Methanol
Flow Rate	0.4 mL/min
Gradient	Time (min) %B 0.0 10 0.5 10 2.0 30 14.0 95 14.5 100
Stop Time	16.5 min
Post Time	6 min
MS Instrument Conditions	
Parameter	*Value*
MS	Agilent 6495 Triple Quadrupole MS/MS Agilent Jet Stream ESI source
Gas Temperature	175 °C
Gas Flow	17 L/min
Nebulizer	20 psi
Sheath Gas Temperature	275 °C
Sheath Gas Flow	11 L/min
Capillary Voltage (Neg)	2500 V
Nozzle Voltage (Neg)	0 V
iFunnel High Pressure RF (Neg) Low Pressure RF (Neg)	 90 V 40 V

Table S4. Precursor and product ions of PFAS: (a) native compounds and (b) corresponding internal standards. Product ion 1 is for quantification and product ion 2 is for confirmation.

(a)

#	PFAS Analyte	Compound Name	Precursor (m/z)	Product 1 (m/z)	Product 2 (m/z)	Isotopically Labeled Internal Standard
1	PFBA	Perfluorobutanoic acid	213	169	-	$^{13}C_4$-PFBA
2	PFPeA	Perfluoropentanoic acid	263	219	-	$^{13}C_5$-PFPeA
3	PFBS	Perfluorobutanesulfonic acid	299	80	99	$^{13}C_3$-PFBS
4	PFHxA	Perfluorohexanoic acid	313	269	119	$^{13}C_5$-PFHxA
5	4:2 FTS	1H,1H,2H,2H-Perfluorohexane sulfonic acid	327	307	81	$^{13}C_2$-4:2FTS
6	PFPeS	Perfluoropentanesulfonic acid	349	80	99	$^{13}C_3$-PFHxS
7	PFHpA	Perfluoroheptanoic acid	363	319	169	$^{13}C_4$-PFHpA
8	PFHxS	Perfluorohexanesulfonic acid	399	80	99	$^{13}C_3$-PFHxS
9	PFOA	Perfluorooctanoic acid	413	369	169	$^{13}C_8$-PFOA
10	6:2 FTS	1H,1H,2H,2H-Perfluorooctane sulfonic acid	427	407	81	$^{13}C_2$-6:2FTS
11	PFHpS	Perfluoroheptanesulfonic acid	449	80	99	$^{13}C_8$-PFOS
12	PFOS	Perfluorooctanesulfonic acid	499	80	99	$^{13}C_8$-PFOS
13	8:2 FTS	1H,1H,2H,2H-Perfluorodecane sulfonic acid	527	507	81	$^{13}C_2$-8:2FTS

(b)

#	PFAS Isotope	Compound Name	Precursor (m/z)	Product 1 (m/z)
1	$^{13}C_4$-PFBA	Perfluoro-n-[1,2,3,4-$^{13}C_4$]butanoic acid	217	172
2	$^{13}C_5$-PFPeA	Perfluoro-n-[1,2,3,4,5-^{13}C5]pentanoic acid	268	223
3	$^{13}C_3$-PFBS	Sodium perfluoro-1-[2,3,4-$^{13}C_3$]butanesulfonate	302	80
4	$^{13}C_5$-PFHxA	Perfluoro-n-[1,2,3,4,6-^{13}C5]hexanoic acid	318	273
5	$^{13}C_2$-4:2FTS	Sodium 1H,1H,2H,2H-perfluoro-1-[1,2-$^{13}C_2$]hexane sulfonate	329	309
6	$^{13}C_4$-PFHpA	Perfluoro-n-[1,2,3,4-$^{13}C_4$]heptanoic acid	367	322
7	$^{13}C_3$-PFHxS	Sodium perfluoro-1-[1,2,3-$^{13}C_3$]hexanesulfonate	402	80
8	$^{13}C_8$-PFOA	Perfluoro-n-[$^{13}C_8$]octanoic acid	421	376
9	$^{13}C_2$-6:2FTS	Sodium 1H,1H,2H,2H-perfluoro-1-[1,2-$^{13}C_2$]-octane sulfonate	429	409
10	$^{13}C_8$-PFOS	Sodium perfluoro-[$^{13}C_8$]octanesulfonate	507	80
11	$^{13}C_2$-8:2FTS	Sodium 1H,1H,2H,2H-perfluoro-1-[1,2-$^{13}C_2$]-decane sulfonate	529	509

Table S5. The kinetic data for (a) PFOA (the initial con. of PFOA ≈ 2 mg/L); QNC 12:1 amount = 0.32 mg (32 mg/L); (b) PFBA (the initial con. of PFBA ≈ 10 mg/L); QNC 12:1 amount = 3.2 mg (320 mg/L); (c) PFOS (the initial con. of PFOS ≈ 5 mg/L); QNC 12:1 amount = 0.32 mg (32 mg/L); (d) PFBS (the initial con. of PFBS ≈ 10 mg/L); QNC 12:1 amount = 3.2 mg (320 mg/L).

(a)

Time	**Batch 1 C_e (mg/L)**	**Batch 2 C_e (mg/L)**	**Average C_e (mg/L)**	**Q_e (mg/g)**	**Error Bar for Q_e (mg/g)**
1 min	0.98	0.80	0.89	34.09	3.34
2 min	0.76	0.71	0.74	39.71	0.77
3 min	0.67	0.60	0.63	43.38	1.19
5 min	0.53	0.52	0.52	47.44	0.23
15 min	0.42	0.41	0.42	51.34	0.19
1 h	0.39	0.39	0.39	52.49	0.05
2 h	0.34	0.29	0.32	55.10	0.77
4 h	0.32	0.27	0.30	55.81	0.78
9 h	0.25	0.28	0.27	56.80	0.57
24 h	0.23	0.25	0.24	57.96	0.34

(b)

Time	**Batch 1 C_e (mg/L)**	**Batch 2 C_e (mg/L)**	**Average C_e (mg/L)**	**Q_e (mg/g)**	**Error Bar for Q_e (mg/g)**
1 min	3.93	3.68	3.81	20.04	0.39
3 min	4.81	3.81	4.31	18.47	1.56
5 min	4.43	3.81	4.12	19.06	0.97
15 min	4.61	4.48	4.54	17.74	0.20
1 h	4.07	3.98	4.02	19.36	0.15
2 h	4.66	4.28	4.47	17.97	0.58
4 h	4.46	4.13	4.30	18.51	0.52
9 h	4.38	4.26	4.32	18.44	0.19
24 h	4.41	4.27	4.34	18.38	0.22

(c)

Time	Batch 1 C_e (mg/L)	Batch 2 C_e (mg/L)	Average C_e (mg/L)	Q_e (mg/g)	Error Bar for Q_e (mg/g)
1 min	0.085	0.037	0.061	185.59	1.91
3 min	0.036	0.034	0.035	186.41	1.09
5 min	0.052	0.063	0.057	185.71	1.79
15 min	0.028	0.050	0.039	186.29	1.21
1 h	0.015	0.017	0.016	187.00	0.50
2 h	0.008	0.016	0.012	187.11	0.39
6 h	0.006	0.006	0.006	187.31	0.19
24 h	0.012	0.014	0.013	187.10	0.40

(d)

Time	Batch 1 C_e (mg/L)	Batch 2 C_e (mg/L)	Average C_e (mg/L)	Q_e (mg/g)	Error Bar for Q_e (mg/g)
1 min	3.68	3.53	3.60	20.11	0.23
3 min	3.18	3.02	3.10	21.68	0.25
5 min	3.01	3.08	3.05	21.86	0.10
15 min	2.96	2.92	2.94	22.19	0.07
1 h	3.10	2.74	2.92	22.24	0.57
2 h	3.54	3.54	3.54	20.31	0.00
4 h	3.07	2.83	2.95	22.15	0.38
9 h	2.85	2.96	2.90	22.30	0.18
24 h	3.01	3.30	3.16	21.51	0.46

Table S6. The isotherm data for (a) PFOA (Initial con. of PFOA ≈ 1-50 mg/L); QNC 12:1 amount = 0.32 mg (32 mg/L); equilibrium time = 24 hrs; (b) PFBA (Initial con. of PFBA ≈ 1-100 mg/L; QNC 12:1 amount = 3.2 mg (320 mg/L); equilibrium time 24 hrs; (c) PFOS (Initial con. of PFBS ≈ 1-50 mg/L; QNC 12:1 amount = 0.32 mg (32 mg/L); equilibrium time 24 hrs; PFBS (Initial con. of PFBS ≈ 1-250 mg/L; QNC 12:1 amount = 3.2 mg (320 mg/L); equilibrium time = 24 hrs.

(a)

Initial Concentration C_o (mg/L)	**Batch 1 C_e (mg/L)**	**Batch 2 C_e (mg/L)**	**Average C_e (mg/L)**	**Q_e (mg/g)**	**Error bar for Q_e (mg/g)**
1.01	0.24	0.21	0.22	28.74	0.61
2.91	0.35	0.55	0.45	90.28	3.80
5.65	0.54	0.39	0.46	190.27	2.65
8.58	0.37	0.37	0.37	301.23	0.07
10.71	0.99	0.83	0.91	359.38	2.99
16.48	5.81	6.46	6.13	379.54	12.02
21.86	9.64	10.23	9.93	380.21	9.37
22.09	12.50	11.93	12.21	362.20	10.43
27.78	17.07	16.68	16.88	399.75	7.07
44.09	32.42	31.84	32.13	388.83	9.40

(b)

Initial Concentration C_o (mg/L)	**Batch 1 C_e (mg/L)**	**Batch 2 C_e (mg/L)**	**Average C_e (mg/L)**	**Q_e (mg/g)**	**Error bar for Q_e (mg/g)**
6.44	3.52	3.08	3.30	9.79	0.69
12.45	5.62	6.34	5.98	20.22	1.13
16.56	5.94	5.61	5.78	33.70	0.52
22.95	8.25	7.72	7.99	46.77	0.82
34.67	10.94	9.95	10.44	75.72	1.54
60.32	28.95	29.14	29.05	97.74	0.29
73.09	39.68	38.60	39.14	106.11	1.68
89.39	55.21	61.17	58.19	97.49	9.31
118.27	88.96	88.89	88.92	91.71	0.12

(c)

Initial Concentration C_o (mg/L)	**Batch 1 C_e (mg/L)**	**Batch 2 C_e (mg/L)**	**Average C_e (mg/L)**	**Q_e (mg/g)**	**Error bar for Q_e (mg/g)**
1.34	0.07	0.25	0.16	37.01	2.75
3.32	0.11	0.15	0.13	99.49	0.55
5.93	0.08	0.09	0.09	182.69	0.05
8.63	0.06	0.07	0.07	267.64	0.06
10.93	0.16	0.36	0.26	333.59	3.18
17.10	0.97	0.98	0.98	503.83	0.16
20.33	3.27	2.76	3.02	551.84	8.23
29.39	11.53	12.19	11.86	564.28	10.64
42.85	26.12	25.96	26.04	551.53	2.69
45.69	29.89	28.40	29.14	542.84	24.34

(d)

Initial Concentration C_o (mg/L)	**Batch 1 C_e (mg/L)**	**Batch 2 C_e (mg/L)**	**Average C_e (mg/L)**	**Q_e (mg/g)**	**Error bar for Q_e (mg/g)**
5.83	1.69	1.54	1.62	13.16	0.24
11.87	3.23	3.04	3.14	27.30	0.30
16.20	4.45	4.18	4.32	37.14	0.42
24.38	6.08	5.65	5.87	57.87	0.67
33.13	7.63	8.36	8.00	78.54	1.13
57.96	13.43	13.68	13.56	138.77	0.39
84.63	28.60	30.23	29.41	172.54	2.54
122.48	52.17	47.88	50.02	226.44	6.70
156.00	86.69	77.34	82.01	231.21	14.61
195.49	109.78	97.46	103.62	287.11	19.24
243.39	157.48	167.84	162.66	258.60	16.59

Table S7. The displacement study for (a) PFBS/PFOS system (the initial con of PFAS ≈ 60 mg/L; QNC 12:1 amount = 0.16 mg (160 mg/L)). PFBS added to the adsorbent, which was equilibrated for 24 hrs. PFOS was added at time = 0 min; (b) PFBA/PFOA system (the initial con of PFAS ≈ 60 mg/L; QNC 12:1 amount = 0.16 mg (160 mg/L); PFBA added to the adsorbent, which was equilibrated for 24 hrs. PFOA was added at time = 0 min.

(a)

Time	C_e of PFBS (mg/L)	Q_e (mg/g)	Deviation	C_e of PFOS (mg/L)	Q_e (mg/g)	Deviation
0 min	28.21	203.88	1.51	51.65	0	0
1 min	49.20	72.69	6.72	2.62	306.43	2.48
3 min	48.78	75.31	17.12	1.95	310.63	1.97
5 min	50.00	67.71	7.56	1.98	310.47	0.85
15 min	50.46	64.82	1.57	1.26	314.95	0.62
1 h	50.47	64.73	2.02	0.79	317.87	0.82
2 h	49.49	70.84	3.07	0.80	317.81	0.59
4 h	50.58	64.08	4.29	0.92	317.07	0.18
24 h	49.34	71.78	3.36	1.02	316.46	0.25

(b)

Time	C_e of PFBS (mg/L)	Q_e (mg/g)	Deviation	C_e of PFOS (mg/L)	Q_e (mg/g)	Deviation
0 s	44.71	128.07	6.18	71.40	0	0
30 s	54.54	66.65	1.02	30.56	255.27	10.14
60 s	60.53	29.17	0	28.18	270.13	5.11
90 s	53.38	73.90	6.83	24.21	294.91	18.44
10 min	54.63	66.07	0.67	20.94	315.40	5.90
1 h	55.40	61.23	3.87	18.92	328.00	1.85
6 h	57.54	47.87	2.16	18.01	333.71	6.18
24 h	56.44	54.75	1.92	17.97	333.91	14.72

Table S8. The competitive study for (a) PFBS/PFOS system (the initial con of PFAS ≈ 60 mg/L; QNC 12:1 amount = 0.16 mg (160 mg/L)). Single-solute means adsorbent was mixed with PFBS or PFOS individually. Bi-solute means PFBS and PFOS were added together to the adsorbent; (b) PFBA/PFOA system (the initial con of PFAA ≈ 60 mg/L; QNC 12:1 amount = 0.16 mg (160 mg/L)). Single-solute means adsorbent was mixed with PFBA or PFOA individually. Bi-solute means PFBA and PFOA were added together to the adsorbent.

(a)

Time	**C_e of PFBS (mg/L)**	**Q_e (mg/g)**	**C_e of PFOS (mg/L)**	**Q_e (mg/g)**
Single-solute				
30 s	34.86	130.26	0.83	300.45
60 s	33.80	136.86	0.83	300.47
90 s	32.14	147.26	0.75	300.92
10 min	31.91	148.70	0.51	302.46
1 h	30.73	156.09	1.94	293.53
6 h	33.02	141.73	0.26	304.02
24 h	29.62	163.00	0.16	304.64
Bi-solute				
30 s	52.67	18.91	2.73	288.57
60 s	49.71	37.46	1.67	295.18
90 s	49.03	41.66	0.98	299.52
10 min	50.87	30.19	0.71	301.20
1 h	50.61	31.84	0.38	303.28
6 h	47.80	49.38	0.19	304.47
24 h	49.40	39.39	0.26	303.97

(b)

Time	C_e of PFBS (mg/L)	Q_e (mg/g)	C_e of PFOS (mg/L)	Q_e (mg/g)
Single-solute				
30 s	45.14	125.38	17.99	333.82
60 s	38.84	164.72	18.48	330.73
90 s	47.25	112.17	18.55	330.33
10 min	41.81	146.22	14.74	354.11
1 h	48.11	106.82	13.42	362.37
6 h	43.47	135.80	9.30	388.12
24 h	43.37	136.42	10.20	382.48
Bi-solute				
30 s	52.25	80.96	26.15	282.83
60 s	53.16	75.24	20.94	315.39
90 s	60.06	32.09	22.15	307.82
10 min	53.18	75.10	16.72	341.76
1 h	55.69	59.46	14.72	354.23
6 h	57.85	45.97	11.45	374.70
24 h	53.78	71.35	12.40	368.77

Table S9. Adsorption experiment with 1 mM and 0.1M NaCl ion (PFAS concentrations: 2 mg/L for PFOS/PFOA with 32 mg/L QNC; 10 mg/L for PFBS/PFBA with 320 mg/L QNC

	PFOS	**PFOA**	**PFBS**	**PFBA**
C_o (mg/L)	1.42	2.11	9.88	9
C_e (mg/L)	0.01	0.02	1.56	2.75
Q_e (mg/g)	46.94	69.42	27.71	20.82
C_e 1mM NaCl	0.01	0.29	6.22	6.2
Q_e 1mM NaCl	46.94	39.67	8.67	3.57
C_e 0.1 M NaCl	0.02	1.43	10.22	10.44
Q_e 0.1 M NaCl	46.61	22.49	0	0

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